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Non-volatile resistive switching device harnessing recombinant protein as reversible metal chelator

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Abstract

A high-performance biomaterial-based resistive switching device is implemented by integrating a thermally denatured protein (hexa-His-tagged recombinant molecular chaperone DnaJ (rDnaJ)) with controllable culpric metal ion chelating properties. The conductivity of the heat-denatured rDnaJ protein layer between the metal electrodes can be reversibly controlled to enable the formation/rupture of conductive Cu filaments by tailoring the metal chelating properties of the amino acid residues in the insulating protein matrix in a pH- and/or redox potential-dependent manner, giving rise to high-performance non-volatile RS behavior.

1. Introduction

There is a growing demand for biomaterials that can be used as the functional components of future devices. This demand is motivated by the need to fabricate building blocks suitable for next-generation biocompatible electronic/photonic devices for a variety of applications. One of the most promising applications of biomaterials in future electronic devices is in resistive switching (RS) devices, used in next-generation information storage/processing devices, wherein the biomaterial acts as the main switching layer with a resistance that is modulated by an electrical stimulus. Recently, several groups have demonstrated the fabrication of RS devices using a variety of biomaterials [1, 2]. Although these studies have demonstrated the feasibility and potential advantages of protein-based RS devices, the device performances must be further improved to enable wide usage in new functional memory and electronic circuits. To satisfy the requirements of a sustainable non-volatile memory and switching device, comprehensive and precise control over the formation and rupture of conductive nano-filaments embedded in the biomaterials using novel pathways is critically required.

2. Experiment

Preparation of the rDnaJ-Coated Substrates

The rDnaJ-coated Pt electrodes were prepared by formulating rDnaJ solutions in 10 mM phosphate buffer at pH 5, 6, 7, or 8 to give a predetermined protein concentration in the range of 1 ~ 4 μM. Subsequently, 20 μL of the rDnaJ solution was drop-cast onto the Pt patterned rigid or flexible substrates using a micropipette. All samples were heated at 70°C for 20 min to induce assembly of a uniform thin denatured rDnaJ protein layer on the Pt electrodes.

Device fabrication

The device structure consisted of a top Cu active electrode, an rDnaJ resistive switching layer, and a bottom Pt electrode on the substrate, as illustrated schematically in Fig. 1. The resistive switching devices were fabricated on a 285 nm SiO2/Si substrate. Pt bottom electrodes were evaporated through a shadow mask. Following rDnaJ deposition, 40 nm Cu top electrodes were deposited through a shadow mask in such a way as to be oriented perpendicular to the Pt bottom electrodes.

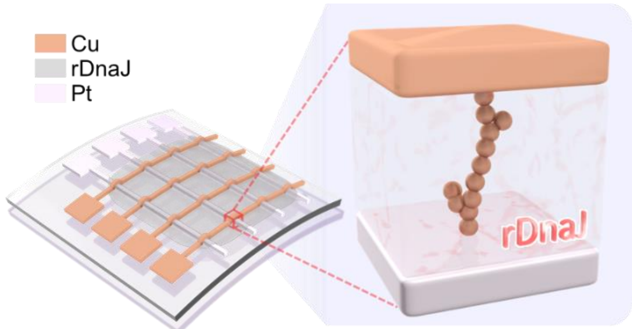

Fig. 1 Schematic diagrams of fabricated rDnaJ RS device array and enlarged single RS devices with the conductive filament.

3. Results and Discussion

The protein RS devices were electrically characterized in the DC voltage sweeping mode over repeated operation cycles (Fig. 2a). The DC bias was applied to the top Cu electrode while the bottom Pt electrode was grounded. Initially, the as-prepared device was in a high resistance state (HRS). As the voltage was swept from zero to positive values with current compliance, the current abruptly increased once the bias reached a threshold voltage of 0.12 V. This is called the SET process, and the corresponding voltage is defined as the set voltage (V_{set}). After the SET process, the device remained in a low resistance state (LRS). As the voltage was swept from zero to negative values, the resistance state changed from the LRS to the HRS at a voltage of −0.08 V, defined as the reset voltage (V_{reset}). This is called the RESET process. As shown in Fig. 2a, the fabricated rDnaJ device displayed stable bipolar RS properties over multiple switching cycles while maintaining extremely low V_{set} and V_{reset} values.

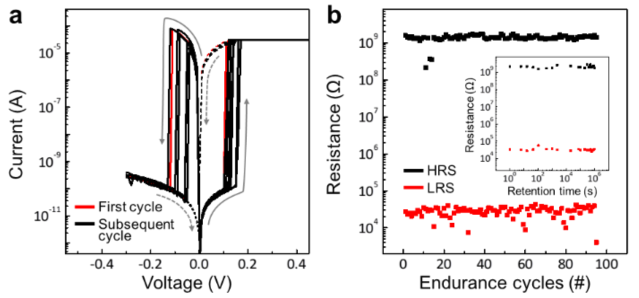

Fig. 2 (a) I-V Characteristics of rDnaJ resistive switching devices. (b) The endurance characteristics for the cyclic set/reset process up to 100 times. The inset shows data retention performance up to 10^6s

Cyclic endurance and data retention tests were carried out to further investigate the stability of the rDnaJ protein RS devices (Fig. 2b). A cyclic voltage was applied from 0 to 0.5 V and then from 0 to -0.3 V, and a read voltage of 0.03 V was employed to avoid a resistance state change. The rDnaJ RS device revealed that the LRS and HRS states remained stable over approximately 100 repeated cycling tests and a test period of 10^6 s. The current ratio between the LRS and HRS state is above 10^6. The endurance and retention characteristics of the rDnaJ RS devices were superior to those obtained from previously reported switching devices fabricated using other biomaterials [1,4-6]. Our results revealed that the use of a controlled rDnaJ switching layer provided high-performance non-volatile memory and switching devices.

The non-volatile properties of the rDnaJ RS device could be further utilized to construct a sequential logic circuit (SR latch), a crucial building block in digital information processing systems. Fig. 3a and 3b plot the schematic diagram and operation of an SR latch circuit composed of two rDnaJ RS devices. These results successfully demonstrated the construction of sequential logic circuits using integrated rDnaJ RS devices, revealing that the rDnaJ RS device can form a fundamental building block for future digital electronic systems. In the LRS state, the current of the rDnaJ RS device was found to be inversely proportional to the current compliance. Fig. 3c plots the current compliance-mediated multi-level state characteristics of the rDnaJ RS device under repeated SET/RESET operation. The SET and RESET voltages remained almost constant whereas the LRS resistance increased as the current compliance conditions changed from 1 to 60 μA. The LRS states were completely restored to the HRS state through the RESET process. A LRS multi-level state in which the resistance ratio of ~100 was deterministically implemented by adjusting the current compliance. In addition, flexible devices have attracted increasing attention due to their potential applications in wearable electronic devices and biomedical devices. A flexible switching device based on a biocompatible material would be highly desirable. The rDnaJ RS devices were fabricated on polyethylene terephthalate (PET) transparent and flexible substrates. We measured the resistance of LRS and HRS as a function of the bending radius to investigate the mechanical bending stability of the rDnaJ RS device on a PET substrate. As shown in Fig. 3d, no significant variations in the resistance or ON/OFF ratio were observed above an ON/OFF ratio of 10^6, demonstrating the excellent flexibility of our rDnaJ RS device.

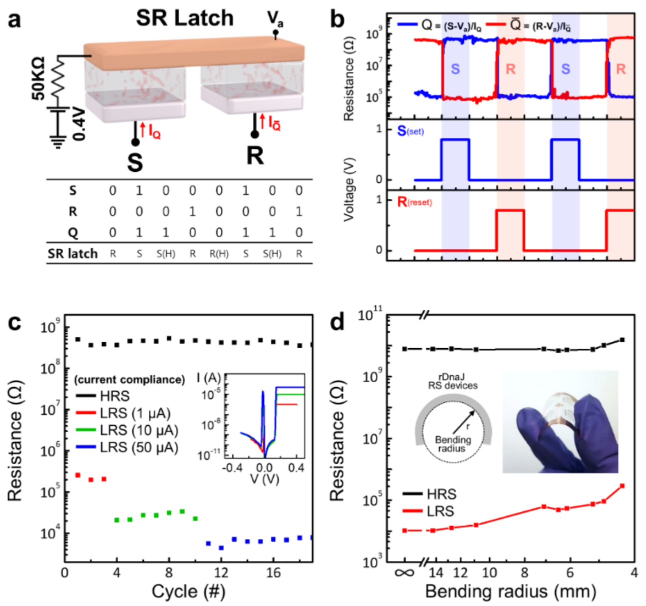

Fig. 3 (a) Schematic diagram of the SR latch and (b) its operating result. (c) Current compliance-mediated multi-level R_{LRS} state characteristics of the rDnaJ RS device. The inset shows each I-V curves. (d) Resistance as a function of bending radius for the rDnaJ RS device fabricated on the flexible PET substrate.

4. Conclusion

In summary, we successfully prepared high-performance biocompatible non-volatile data storage and RS devices by integrating an rDnaJ switching layer, with controllable metal ion chelating properties, into an rDnaJ RS device. The low-power operation and non-volatile RS devices were fabricated by integrating pH-controllable metal chelating sites in the form of the rDnaJ protein. The excellent switching properties and non-volatile characteristics of the rDnaJ RS devices were deployed in SR Latch sequential logic circuits. rDnaJ RS devices were shown to successfully enable multi-level switching operation and the reliable operation of flexible devices. These results show great promise of the rDnaJ RS device as a fundamental building block for various device applications.

References

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